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LOCAL FALLOUT FROM NUCLEAR TEST DETONATIONS (U)

VOLUME IV. ANNOTATED COMPENDIUM OF DATA ON RADIOCHEMICAL AND RADIATION CHARACTERISTICS OF FALLOUT (U)

PART 2. RADIOCHEMICAL COMPOSITION, INDUCED ACTIVITY, GAMMA SPECTRA (U)

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Washington, D.C. 20305

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**VOLUME IV. ANNOTATED COMPENDIUM OF DATA
ON RADIOCHEMICAL AND RADIATION
CHARACTERISTICS OF FALLOUT (U)**
**PART 2. RADIOCHEMICAL COMPOSITION,
INDUCED ACTIVITY, GAMMA SPECTRA (U)**

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ADMINISTRATIVE INFORMATION

NOLTR 72-137

31 May 1972

Local Fallout from Nuclear Test Detonations Volume IV.
Annotated Compendium of Data on Radiochemical and Radiation Characteristics of Fallout Part 2. Radiochemical Composition, Induced Activity, Gamma Spectra

This work has been supported by the Defense Nuclear Agency under Subtask PD013.

ROBERT WILLIAMSON II
Captain, USN
Commander

Albert Lightbody
ALBERT LIGHTBODY
By direction

ACKNOWLEDGEMENTS

The work on this volume was started at the Naval Radiological Defense Laboratory. The closure of that laboratory in 1969 caused a serious interruption in the project. An attempt was made to finish the volume at the Naval Weapons Laboratory, but this did not prove to be feasible. The volume has finally been completed at the Naval Ordnance Laboratory.

Because of the complex history of this project many people were involved in the completion of the work. Inevitably, proper acknowledgements to some of the people have been overlooked and to these we apologize. However, thanks are due to E. C. Freiling for his continued interest and guidance. Special appreciation should be given to H. Goya, for his editorial assistance, and to L. Buggs for typing the copy.

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ABSTRACT

Many technical reports have been published that contain data describing local fallout from nuclear weapon tests. Data on fallout from dissimilar types of weapons detonated under different conditions usually are not directly comparable, and thus far only limited attempts have been made to summarize these data so as to characterize local fallout. Correlating a given body of data would require that it be converted to a standard form, the significant parameters be computed, and these be compared according to some theory or empirical formula. A prerequisite would be the compilation of the data and authoritative commentary. For this purpose, a study was made that resulted in five volumes of such data, published as a series under the general title Local Fallout from Nuclear Test Detonations.

The study was intended to critically select and compile, from published reports, data on all characteristics of local fallout from nuclear weapon tests held through 1958. These reports would have included all pertinent data from tests concluded prior to the first moratorium on above-ground test detonations. However, the Dominic Test Series of 1962 extended the time span to be covered; thus, the first four volumes contained the data for tests through 1958. The last volume of the Series includes material published on weapon tests conducted through 1962.

The first volume in the series, Indexed Bibliography of United States and British Documents on Characteristics of Local Fallout, was published in 1961. It made available a ready reference to 206 reports from which relevant information was to be selected, while the selection and compilation of data progressed. The volume consists of an index to the references by fallout characteristics, an index to the references by authors' names, and a list of the references.

The second volume, Compilation of Fallout Patterns and Related Test Data (U), was published in several parts in 1963-66. It contains information on fallout patterns, on wind and cloud data, and on all that is pertinent to shot conditions, such as yield, burst height and placement.

The third volume, Annotated Compendium of Data on Physical and Chemical Properties of Fallout (U), was published in 1966. It contains the more significant of the data published that pertains to the physical and chemical nature of local fallout particles. Included are general statements made by the original investigators on the physical and chemical nature of the particles, as well as specific observations produced by detailed experimental analyses. Also included wherever possible are data

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descriptive of the samples analyzed, together with relevant information on collection equipment, location of collection, processing involved, and purpose of the analyses.

This, the fourth, volume is a compendium of data on radiation and radiochemical characteristics of local fallout. It is published in two parts. Part 1 presents data on radioactivity investigated as a function of mass, particle size, and time. Accordingly, Part 1 is organized into three main data sections: Specific Activity, Activity-Size Distribution, and Decay. This part, Part 2, presents data relating activity measurements with radiochemical content (sections on Radionuclide Composition and Induced Activity) and energy (section on Gamma Spectra).

Again, as in the third volume, included besides the primary data is background information on the location and types of collection equipment used, the instrumental or radiochemical processing involved, and the purposes of the analyses.

The fifth volume, Transport and Distribution of Local (Early) Fallout From Nuclear Weapon Tests (U), was published in 1965. It presents and evaluates information relating to nuclear detonations through 1962, that concerns the formation of the particle cloud, particle fall behavior, fallout dynamics, and fallout pattern features.

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CLASSIFICATION GUIDANCE

Information in this report is classified as follows:

1. Yields are Confidential-Formerly Restricted Data when less than 1 megaton, Secret-Formerly Restricted Data when equal to or greater than 1 megaton, unless they have been announced by publication in The Effects of Nuclear Weapons, in which case they are Unclassified.

2. Event names not listed in The Effects of Nuclear Weapons are Confidential.

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SECTION I INTRODUCTION

THE SERIES

The study of fallout began with the first nuclear detonation but, in the early years, the magnitude of the explosion effects overshadowed the significance of the radiation effects. When humans were accidentally exposed to radioactive fallout and when fallout generation was examined as a military tactic, the phenomena of fallout - its mechanics and effects - were investigated more thoroughly and more comprehensively.

As nuclear detonations and tests became more sophisticated and highly instrumented, more and more data on fallout was reported. However, such information was not always available to investigators not involved in a test; the participating projects published their detailed findings in separate reports or the information was summarized in final reports, and the reports were classified. Often pertinent details of the collection of samples were neglected or overlooked when such information would have helped explain discrepancies in the data that became apparent when the samples were analyzed.

Fallout was produced and information was collected at an increasing pace during the thirteen years from the first nuclear detonation to 1958, the date of the first moratorium. Again, during the testing in 1962 and until the second moratorium became effective, data on all phases of fallout production, transport, and characteristics were reported, most frequently in classified documents.

Many of the projects of these field tests were conducted independently, with each project doing its own planning to achieve its objectives. Consequently, the vast quantity of data from all these independent experiments has not been examined collectively and has been correlated very little. This probably resulted from the physical scattering of the data and the lack of a complete literature search.

Such a search was made of the reports from these tests, and a compendium of data was prepared as a series of volumes under the general title Local Fallout from Nuclear Test Detonations. Over 200 documents were selected as sources of information and data essential to studies for the definition of the physical and chemical parameters of the local fallout produced by nuclear detonations. These documents are listed in the indexed bibliography that constitutes Volume I of the Series. This compendium is being issued to bring together in one set of documents the significant data on fallout that would be useful for the understanding of the formation of radioactive debris, the physical parameters of the fallout material itself, and the long-range residual effects and hazards associated with the dispersed radioactivity.

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INTRODUCTION
THE SERIES

This is Part 2 of the fourth of the five volumes that comprise the Series.

The other parts of the Series are:

Volume I. F. K. Kawahara, H. Lee. Indexed Bibliography of the United States and British Documents on Characteristics of Local Fallout (U). U. S. Naval Radiological Defense Laboratory, DASA-1251, Vol. I (USNRDL-469), 27 June 1961 (Confidential).

Volume II. M. Morgenthau, H. B. Meieran, R. L. Showers, J. L. Morse. Compilation of Fallout Patterns and Related Test Data (U). U. S. Army Nuclear Defense Laboratory, DASA-1251, Vol. II, Parts 1 and 2 (NDL-TR-34, Parts 1 and 2), August 1963 (Secret-Restricted Data).

M. Morgenthau, R. L. Showers. Supplement. Foreign Nuclear Tests (U). U. S. Army Nuclear Defense Laboratory, DASA-1251, Vol. II, Supplement (NDL-TR-34, Supplement), October 1964 (Secret-Restricted Data).

R. L. Showers, N. J. Dombek, A. G. Garcia. Part 3. Nougat Through Niblick (U). U. S. Army Nuclear Defense Laboratory, DASA-1251, Vol. II, Part 3 (NDL-TR-34, Part 3), March 1966 (Secret-Restricted Data).

Volume III. F. K. Kawahara, J. D. O'Connor, H. Lee, M. A. Connors. Annotated Compendium of Data on Physical and Chemical Properties of Fallout (U). U. S. Naval Radiological Defense Laboratory, DASA-1251, Vol. III (USNRDL-497), 10 November 1966 (Secret-Restricted Data).

Volume IV. J. D. O'Connor, G. R. Crocker. Annotated Compendium of Data on Radiochemical and Radiation Characteristics of Fallout (U). U. S. Naval Radiological Defense Laboratory, DASA-1251, Vol. IV, Part 1 (NRDL-68-2), 11 September 1968 (Secret-Restricted Data).

Volume V. P. D. LaRiviere, S. L. Brown, J. D. Sartor, C. F. Miller. Transport and Distribution of Local (Early) Fallout From Nuclear Weapons Tests (U). Stanford Research Institute, DASA-1251, Vol. V (NDL-TR-65; SRI-4-3338), May 1965 (Secret-Restricted Data).

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INTRODUCTION
THIS VOLUME

THIS VOLUME

Volume IV is a two-part compilation of critically selected data from radioactivity measurements of fallout samples produced by large-scale tests of nuclear weapons by the United States. As with the other volumes of the Series, attention has been restricted to local or close-in fallout.

The nature of the data dealt with in this volume (measurements of radioactivity) required the frequent exercise of critical judgment on the part of the compilers in deciding what portions of a fairly large body of published activity measurements on fallout were worthy of inclusion. It seems appropriate here to review briefly some of the problems involved in measuring fallout radioactivities, the circumstances under which the data were acquired, the techniques employed in making the measurements, the methods followed in reducing and reporting data, and the general criteria adopted by the compilers for selecting data for inclusion in the volume.

SCOPE OF DATA

The data in this volume consist, in the main, of measurements of ionizing-radiation properties in terms of physical or chemical characteristics - the activity-size distribution, the radionuclide content, or the energy spectra, for example - of a fallout sample. These determinations are possible only because of the radioactivity inherent in the true "fallout" part of the sample, as distinguished from the inert debris also disturbed by the event. The radioactivity that poses the hazard also permits definition of the problem and its extent.

There is less descriptive text from the source documents in this volume, in comparison with Volume III, since much of the data is in tabular form (count rate or disintegration rate per unit of time) or as decay curves (decrease in count rate as a function of time) and energy-spectrum diagrams. Explanatory text from the source documents is presented where useful. Commentary by the authors of this volume is limited to that needed to clarify the sampling or analytical techniques or to amplify the resulting data.

CONSIDERATIONS IN SELECTION OF DATA

Radioactivity measurements are intended to assay the rate at which radiation is emitted by unstable nuclides as they decay to more stable states. In the case of fallout, measurements are usually restricted to beta and gamma radiation. The third kind of nuclear radiation (alpha radiation) is always detectable in fallout but is of slight importance, since the penetrating power of alpha rays is low and the levels of alpha

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activity in fallout from devices of appreciable yield are invariably insignificant. As a matter of fact, the products of fission events themselves, with the exception of two nuclides of exceedingly long half-lives do not emit alpha radiation at all. The alpha activity in fallout is due primarily to the presence of unfissioned uranium and/or plutonium isotopes, or to isotopes produced by neutron activation of this material.

Essentially all fission products emit beta radiation at the rate of one beta particle per disintegration.* Gamma radiation, on the other hand, is not emitted at all by many fission products and the number of photons per disintegration from the gamma-emitters varies widely. For instance, ^{85}Kr emits only about 0.01 gamma photons per disintegration, while ^{83}Se emits an average of roughly 4.2 photons (of many different energies) per disintegration. A large proportion of gamma and beta activity measurements are made with "counting" instruments which record individual disintegration events, i.e., beta particles or gamma photons reaching the detector. If it were possible to count all the events in a pure radioactive substance for which the photon or particle abundances per disintegration were known, the counting rate would be an absolute measure of the activity. If a mixture of radioactive substances were present, it would be necessary to measure the total activity repeatedly over an appropriate time interval and then to resolve the decay curve into its proper activity components. This latter procedure would be successful only if all the half-lives of the substances present were sufficiently different.

In practice, the determination of absolute counting rates requires rather elaborate and painstaking procedures, even for pure substances. Many corrections must be carefully accounted for: the geometry of the sample, the length of the path from sample to detector, the thickness of the sample, the energy response characteristics of the detector, the resolving time of the counting circuitry, and such effects as backscattering by the sample mounting material and bremsstrahlung in the detector. Because of the mass of absorbing inert material present, fallout samples, generally speaking, are poorly suited for accurate beta-counting and only slightly better suited for gamma-counting. In addition, the radioactivity of fallout samples is produced by a complex mixture of radioactive substances (the fission products), many of which may have closely similar half-lives. The relative proportions of these substances may vary widely from sample to sample, on account of fractionation phenomena which accompany the process of fallout formation. Thus, there is no possibility, in general, of resolving a fallout decay curve into its individual activity components, even if absolute counting data were obtainable.

*Some metastable states of fission-product radionuclides decay to the ground state without beta emission. If these states are regarded as distinct radionuclides, the statement above does not hold exactly.

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INTRODUCTION SELECTION OF DATA

If absolute beta counting data could be obtained, it would be unnecessary to refer the measurements to other standard measured counting rates or theoretical calculations. Since geometry and absorption factors for fallout samples usually cannot be controlled closely enough to obtain absolute beta counting rates, a reference standard is required to lend significance to the data. One of the procedures which has been used is to calibrate the counter with samples of known amounts of a standard activity, such as RaE, in a well-defined and reproducible geometry. The counting rates for fallout samples are then expressed in terms of "equivalent" amounts of the standard activity. This method is far from satisfactory since, because of differences in energy and source mass and geometry, the "equivalence" of the sample to the standard is largely an illusion.

The reduction of gamma counting data on fallout samples to absolute terms is even more difficult than the reduction of beta counting data, because of the wide variation (mentioned earlier) in the number of gamma photons emitted in the disintegration of different fission product radio-nuclides. For this reason, gamma counting data are almost always regarded simply as relative measurements of activity.

It is apparent that beta- and gamma-counting data or gamma ionization chamber data for fallout, even under favorable circumstances, must be interpreted with considerable care. In general, absolute beta counting rates are directly equivalent to disintegration rates, while absolute gamma counting rates are only roughly proportional to disintegration rates. From this point of view, beta counting might seem preferable for fallout studies, since absolute beta count rates determined by different investigators on different samples (even from different events) should be directly comparable, while gamma count rates from different investigations can be compared only if they are referred to comparable standards. On the other hand, the uncertainties that attend beta counting of fallout samples preclude the possibility of obtaining reliable absolute count rates on most fallout samples, so that reference to a standard is always desirable and usually necessary. Furthermore, the external beta-radiation hazard to personnel from fallout is negligible, compared with the external gamma exposure, so that gamma measurements are of more operational significance. However, the Office of Civil Defense has become concerned with the effects of beta radiation and soft photons on plant and insect life.

It should be mentioned that any observation of a quantitative nature of the intensity of radiation is in a sense a radioactivity measurement. Thus, various kinds of field-meter readings, dosimeter readings, film-darkening measurements, etc., might, under the appropriate circumstances, be regarded as radioactivity measurements. The weapon-test literature contains reports of many such observations, but

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they have not been included in this compilation since they cannot be interpreted except in the specialized context in which they were reported.

The data used for this compilation were acquired during the weapons testing period from its beginning through Operation Hardtack in 1958. It should be recalled that most of this period also represented a developmental phase insofar as instrumentation and techniques for radiation-measurement were concerned. During the early part of the period most radiation measurements on fallout were made with metering devices, either of the Geiger-Mueller counting type or the ionization current type. These devices were subsequently replaced by proportional counters and 4-pi ionization chambers, and the significance of some of the radiation measurements reported by investigators improved. In the last years of the period, sodium iodide (NaI) scintillation detectors came into general use for gamma counting. No important use was made of gamma pulse-height analysis instrumentation until after Operation Hardtack.

It is worth remembering that much of the data were obtained under difficult and trying conditions. Personnel and equipment often had to be transported long distances into localities with poor facilities for living and working and with climates unfavorable to men and instruments. The laboratories involved in fallout studies during the period also contended with chronic shortages of equipment and qualified personnel. It would be unrealistic to suppose that these factors did not at times affect the quality of the measurements that were made and reported.

PRESENTATION OF DATA

The data presented are in the form of discrete extracts taken from the various source documents: passages of text, whole or portions of tables, and illustrations. The extracts are verbatim copies except where fragmentary corrective or explanatory information is inserted, within square brackets. A boxed entry identifies the document and page on which the original data appears. With most extracts, the shot to which the data refers is identified within brackets near the beginning of the extract. The extracted data, and the commentary by the present authors, are presented in the Compendium of Data, Section II.

The extracts and related commentaries are organized according to a scheme of classification of the data they describe.* Specific data is classified under detailed categories as follows:

1. First, according to the indicated characteristic; actually, all of the data can be considered radioactivity investigated as a function of:
 - a. mass (specific activity)
 - b. particle size (activity-size distribution)
 - c. time (decay)

*Part 1 is comprised of items a, b, and c. Part 2 is comprised of d and e.

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INTRODUCTION
PRESENTATION OF DATA

- d. radiochemical content (radionuclide composition and induced activity)
- e. energy (gamma spectra)

2. Second, the data under each of the above characteristics are classified further according to location of burst in the environment: air or balloon (all devices detonated well above the earth's surface - dropped from air-craft, shot from guns, lofted by rockets, or suspended by balloon); tower; ground surface (on or close to the land surface); underground; water surface (on or close to the water surface, usually on barges); underwater.
3. Last, the data fitting any combination of a first and second category is subdivided according to the nuclear weapon test operation that produced the data, in this chronological order:

Trinity
Crossroads
Sandstone
Ranger
Greenhouse
Buster-Jangle
Tumbler-Snapper
Ivy
Upshot-Knothole
Castle
Wigwam
Teapot
Redwing
Plumbbob
Hardtack I
Hardtack II

Besides this classification by individual operation, broader categories refer to more than one operation. As indicated in Volume I, some of the source documents generalize, summarize, or compare data according to qualities common to several operations: tests over a span of years, height of detonation, detonation environment. These categories are placed in the sequence of operation names according to the nearest point in time. Where the document does not also mention the specific operations involved, data could not be included under those operation names.

The data is organized for presentation in unit sections in the Compendium of Data, Section II. Each unit section contains data relevant to one combination of the three categories. This organization of data is indicated by the running heads at the top of each page. Each running head is a combination of three terms representing the three categories.

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Particular information can be found under one or another of the various combinations of category terms. For example: radionuclide composition data from all air shots in Operation Ranger can be found under

Radionuclide Composition
Air or Balloon, Ranger

The Table of Contents also assists the reader to locate data pertinent to his particular interest.

Each unit section (all pages with a common running head) begins with a display of introductory information, and the first page of the unit sections can be identified by the consistent format used. It gives the date and location of the weapon test. It lists the source documents for the included data. It lists the relevant shots of the operation, each with its yield, and height or depth if not a surface detonation. The numbers and letters within parentheses following a shot name are additional designations of the shot and indicate its sequential order in the operation. (More detailed information on the device for the shots listed can be found in Volume II of this series.) In each unit section, the commentary by the authors of this volume follows this introductory information. For each document listed, when possible, general information is given on where the subject sample was collected, how it was collected, how it was processed, and, where possible, why the data were collected. Occasionally, the commentary abstracts textual statements from a source document for which no associated data could be extracted and presented.

The introductory information and the commentary at the head of a unit section are extracted from source documents that contain information on the same shots, one or more. However, the extracts from the documents - which occupy the remainder of a unit section - are in numerical order of reference and page numbers; except for variations for convenience of presentation.

Although it was intended to include data in a unit section that are pertinent only to the running head shown, some extracts show other information. Such extracts were reproduced in toto to avoid the work of separating the data not immediately relevant, which did not seem worthwhile.

Throughout the Compendium of Data Section, the source documents are keyed by reference number and first author, to the List of References, Section III. This list of references is based on that included in Volume I of this Series. However, in the present list British documents have been excluded, only those references relevant to Part 2 of this volume are given, and some bibliographical details were up-dated.

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INTRODUCTION DISCUSSION OF DATA

GENERAL DISCUSSION OF DATA

Volume I of the Series, based on the results of the initial literature survey, assigns groups of data to the various categories of Volume IV. However, further, more detailed examination has revealed that many of these assignments would be more appropriately made to other categories of Volume IV, or occasionally even to a different volume. These changes are noted in unit sections for the categories designated in Volume I.

Radionuclide Composition

From the inception of nuclear weapons testing, radiochemical studies of the debris have played a role of fundamental importance. At the outset of the testing program interest in the radiochemical composition of debris per se was limited, since the tactical importance of fallout and its residual radiation properties was somewhat obscured by the more spectacular blast, shock, thermal and initial radiation effects. Nonetheless, the development and improvement of nuclear weaponry was intimately dependent upon radiochemical studies. Much of the phenomenology of a nuclear explosion cannot be studied on a laboratory scale and studies of the fission products in the debris provided the a posteriori evidence for elucidating many of the details of the nuclear reactions in the device. The importance of radiochemical investigations of this kind, often called diagnostic radiochemistry, has not diminished; and with the recognition of the importance of fallout, the need for a complete and detailed understanding of the distribution of fission products in the debris was established. It should be noted that diagnostic studies and fallout studies are not exactly equivalent since certain radioactive products may be very sensitive diagnostic indicators but completely insignificant as far as the residual radiation properties of the debris are concerned. Conversely, extremely important contributors to residual radiation may be diagnostically uninteresting.

The radioactive constituents of nuclear debris include fission products and various induced activities resulting from nuclear reactions on the elements present in the device and the environment. The fission products are the most important and they have therefore received by far the majority of study and documentation. However, the induced activities can also make large contributions to the gamma exposure rate under a number of circumstances and they also demand consideration.

The radiochemical methods used for analysis of fallout samples are well-standardized and except for advances in the separation chemistry of the lanthanides and actinides, they have not changed much since the first tests. The sample of fallout is dissolved along

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with added carriers or isotopic tracers for the fission products, the latter are separated individually from the solution, purified, and assayed for radioactivity by counting. These methods are tedious and time-consuming and require great care on the part of the analyst in order to yield reliable results. The very rapid gamma spectrographic methods of analysis, which are widely used today and remove much of the human element from the analyses, were not sufficiently developed to be of much use during the period covered by this volume.

One of the most formidable problems in fission-product analysis of nuclear debris is the reduction of the data to absolute amounts of the fission products present in the sample. This can be accomplished only by painstaking standardization of procedures, in conjunction with rather elaborate instrument calibration based on standard samples of known assay. In order to compare results from two or more different laboratories, it is necessary to establish that the laboratories are agreed on calibration or, in the absence of agreement, what the magnitudes of the differences are. During the testing period the several laboratories chiefly involved in the radiochemistry of nuclear debris developed in intensive program of establishing and maintaining a state of reasonable intercalibration.

The calibration and intercalibration problems are reflected, to some extent, in the choice of the units for reporting fission-product radionuclide concentrations in nuclear debris. Perhaps the most convenient unit, at least for fallout studies, is equivalent fissions. The number of equivalent fissions of any nuclide in a sample is the number of uranium or plutonium atoms which must have undergone fission to produce the amount of the nuclide found. However, the use of the equivalent-fissions unit depends upon knowledge of the production ratios for the fission process in question. In the case of nuclear weapons these ratios frequently were not known with certainty in advance (it was often one of the aims of the radiochemical studies to determine them) and, accordingly, the unit was not generally used. An exception is the case of ^{99}Mo for which results are frequently expressed as equivalent fissions or, by abbreviation, as "fissions". It was recognized rather early that the yield of ^{99}Mo does not vary much for the types of uranium and plutonium fission of interest - it is always around 6%. Accordingly, ^{99}Mo was often used as the primary indicator of "the fraction of the bomb represented by the sample".

Several other fission-products were reported in equivalent fissions for the first nuclear test, Trinity. The production ratios utilized in this case were derived from certain in-pile fission experiments on plutonium. Subsequently, it became general practice to use the well-known production ratios for thermal-neutron fission of ^{235}U as a standard with which to compare the ratios found in weapons debris.

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Ultimately, a double ratio known as an "R-value" came into general use. This quantity can be better symbolized by $R^{99}(i)$, and is defined:

$$R^{99}(i) = \left[\frac{a_i}{a_{99}} \right] \text{ debris} / \left[\frac{a_i}{a_{99}} \right] U^{235}, \text{ thermal n}$$

Here, a_i and a_{99} represent the number of atoms of nuclide i and ^{99}Mo , respectively. It is important to note that the ratio in the denominator is not merely taken from the literature but is determined experimentally, using the same procedures and instruments used to determine the ratio in the numerator. Instrumental and procedural influences on the absolute magnitudes of the concentrations thus tend to cancel out and $R^{99}(i)$ -values determined by different laboratories on the same sample should agree closely.

The majority of the extracts presented in this volume report results as $R^{99}(i)$ -values, generally designated simply "R-values". Many of the tables appear, at first glance, to be listing simple ratios such as $^{89}\text{Sr}/^{99}\text{Mo}$, etc., but inspection of the table headings reveals the ratios are really the $R^{99}(89)$, etc., double ratios. The R-values are usually near unity for nuclides like ^{95}Zr and ^{140}Ba , which lie in the peak regions of the chain-yield distribution curve for fission. The R-values of nuclides on the wings and in the valley of the curve may show large departures from unity.

Units other than R-values are occasionally used in the extracts. These are generally absolute activities referred to some fiducial time-point, or ratios of such activities, and are either self-explanatory or accompanied by explanatory material.

In addition to the fission products, other radioactivities produced by the action of neutrons on device and environmental materials are found in nuclear debris. These induced activities sometimes assume major significance, both to diagnostic studies and fallout studies. Many of the reports dealing with fission-product radiochemistry also contain some data on induced activities. Although this volume contains a section devoted to induced activity, it appeared impractical in many cases to separate fission-product data from induced-activity data, inasmuch as the original authors of the reports made little distinction in their treatment of the two. Accordingly, the Induced Activity section of this volume contains only extracts that deal exclusively with induced activities. Extracts which treat fission-product activities and induced activities jointly are to be found in Radionuclide Composition section.

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Some of the most formidable problems in nuclear debris radiochemistry result from the phenomenon of fractionation. The fission products are produced by fission processes in fixed proportions, but the proportions found in debris are often far from the expected values and may vary considerably from sample to sample. This failure of the representative proportions to evidence themselves in individual samples is due to differences in the condensability of the elements composing the fission-product decay chains during the rapid quenching of the fireball. As a result, chains which contain rare gases and halogens (volatile) chains, e.g., mass 89 and mass 137) may condense at a different time and in a different way from chains which do not contain these elements (refractory chains, e.g. mass 95 and mass 144). The fractionation effect seems to be present in nearly all samples from all types of testing events, although the degree of severity of the effect varies. The user of the data extracts in this volume will find abundant evidence of fractionation, as well as frequent reference to the phenomenon in the extracted discussions and interpretations of the data.

It is the opinion of the compilers that the radiochemical data contained in this volume differ widely in quality, but no attempt has been made to assign limits of reliability to any parts of it. For the most part the original reports either do not discuss reliability or discuss it in a very perfunctory way. It is customary in radiochemical analysis to assign random error limits (usually rather small) to reported values, but these cannot be taken as literally as similar limits in other kinds of chemical analysis. The determination of tracer quantities of radioactivities is sufficiently complicated to provide many opportunities for matters to go badly astray. Unfortunately, the nuclear debris analyst will frequently have no way of knowing that a certain result is much too high or much too low. Consequently, many determinations which in more conventional circumstances would have been discarded as unacceptable, have found their way into the reports. We know of no satisfactory way of weeding out these pieces of misinformation.

Induced Activity

The presentation of material in this section will deviate slightly from the format of the other sections of this Volume and of Volume III of the Series.

The format that had been followed is to list the references and the detonations covered by the extracted data at the beginning of a new section. Each reference is discussed separately and the project objectives, collection of samples, specialized techniques, and the physical measurement or chemical analysis that was conducted are mentioned or described. In many previous instances, the project description did not

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change but the discussion of techniques or measurements was tailored to suit both the samples analyzed and the radiochemical parameters determined.

Since the sections labeled Radionuclide Composition and Induced Activity both present the results of individual nuclide analysis, and these sections follow each other so closely, the introductory discussion has been curtailed in the Induced Activity unit. This was done to reduce repetition of information since the same samples or analyses usually produced both types of data.

In the presentations in which nothing was added or changed in this introductory material, the reader will be advised to consult the corresponding section in the section, Radionuclide Composition, for details.

Gamma Spectra*

The unique hazards associated with fallout from nuclear explosions are, of course, the direct result of the radiations emitted by the fission-product and other radionuclides incorporated in the fallout particles. With the gradual recognition, during the early phase of nuclear weapons testing, of the potential military significance of fallout, came the recognition of the need for knowing the flux and energy distribution of the radiation. This sort of quantitative description is a necessary preliminary to accurate calculation of such practical matters as exposure hazards and shielding protection factors.

If sufficiently accurate knowledge of the composition of fallout with respect to the radionuclides had been available, along with a knowledge of the decay and radiation properties of the radionuclides, the gamma and beta spectra could have been calculated. In the earlier phases of nuclear testing such calculations could not be reliably made and spectral information from field studies was very important. Even when such calculations became feasible (within the past decade) the complexity and the uncertainties involved made experimental verification highly desirable.** At best such calculations can be made only for unfractionated fallout products and unfractionated compositions probably never occur in the field. The state of fallout-formation theory does not yet permit accurate prediction of the compositional parameters for

*The section heading "Gamma Spectra", which was selected by the compilers of Volume I and has been used in other volumes of this series for cross-referencing purposes, has been retained in this volume to avoid confusion. The section actually contains data on both gamma and beta spectra.

**The latest calculations and the latest direct experimental measurements of fission-product gamma spectra for unfractionated fission product mixtures are not yet in completely satisfactory agreement.

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Fission and fusion processes themselves release tremendous amounts of radiative energy. However, this radiation (usually referred to as "prompt" radiation) cannot be regarded as a property of fallout and, accordingly, is not dealt with in this volume. The radiations from fallout (usually called "residual" or "delayed" radiation) are the gamma photon*, beta particle, alpha particle, and neutron emissions which accompany the decay of the fission products and other radionuclides. Of these, the alpha and neutron radiations occur to so slight an extent that they have customarily been disregarded in considerations of fallout radiation properties. They will not be further discussed here.

The hazard from exposure to beta radiation from fallout has generally been considered to be of a much lower order than exposure to the corresponding gamma radiation. Nevertheless, certain observations, such as the beta burns suffered by the Marshallese Islanders during exposure to fallout from Operation Castle, the known effects of the lodgement of the beta-emitter ^{90}Sr in bone tissue, and speculative effect of short-range beta-radiation absorption by plant and animal life, have brought a certain amount of continuing effort to bear on the problem of determining the distribution of beta energies in fallout. The beta spectrum of fallout is known, from theoretical considerations, to be continuous and rather broad. Direct spectrometer measurements of the shape of the beta spectra of fallout samples are evidently impractical, on account of the difficulties involved in controlling or correcting for the absorption and scattering processes that take place in the fallout sample. No attempts at such measurements were encountered in this survey of the weapons test literature. However, many efforts have been made to resolve the spectra into two or three "effective" components, by the analysis of beta-absorption curves or depth-dose curves. In spite of obvious shortcomings, the results of such studies have been extracted and included here in default of better information.

In the earlier phases of weapons testing, spectrometer measurements of gamma radiation were also impractical. Here again, the resolution of absorption curves into "effective" components was resorted to in an effort to gain some rough idea of the energy distribution of the gamma radiation. However, during the period covered by this volume, spectacular advances were made in the field of gamma-ray spectrometry. Scintillation spectrometers with large NaI(Tl) crystals became generally available along with satisfactory photomultiplier tubes, for handling the light output on the crystals, and multi-channel analyzers for accumulating pulse-height data. From Operation Castle onward reasonably reliable gamma-emission spectral data obtained with this kind of instrumentation began to appear in the weapons test literature. In view of

*"Gamma photon" and "gamma radiation", as used in this discussion, include X-ray photons and X radiation.

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the availability of such information and the limited usefulness of the gamma-absorption curve data, the results of studies of the latter nature have not been included in this volume.

In several of the studies of gamma radiation encountered, the instrumentation was placed in the fallout field for the accumulation of data, in contrast to the alternative method of bringing fallout samples into the laboratory to make the measurements. The field situation is not very satisfactory for the determination of gamma-emission spectra from fallout for the following reasons: (1) In the field large amounts of degraded or scattered low-energy radiation reach the detector, and there is no way of separating this component from the unscattered direct component of the radiation; and (2) the radiation field is generally anisotropic; i.e., the response of the detector changes with its orientation to the field. As a result, the data from the field studies are apt to be misleadingly different than those from laboratory studies. We have, accordingly, not generally included extracts of measurements of this kind. However, the references containing reports of field studies of gamma radiation are mentioned and the reader with specialized interest in such work is referred to the original documents.

The period covered by the material in this volume ended in 1958. Since that time the field of gamma spectrometry has continued to advance with almost explosive rapidity. In particular, the semi-conductor detector with its vastly improved resolution has come into general use along with multiparameter analysis and greatly improved coincidence detection. Furthermore, the use of very sophisticated data-reduction techniques dependent on the availability of large-capacity high-speed computers has become common. Gamma-spectral studies which seemed very significant at Operation Hardtack and prior tests may now have the appearance of being unsophisticated at best and crude or naive at worst. The compilers of this volume have, indeed, been hard-pressed to determine whether most of the work was of anything other than historical interest. We have tried to err on the side of conservatism and hope that nothing of permanent interest has been omitted.

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SECTION II COMPENDIUM OF DATA

The following section contains all the reported data selected as relevant, and useful, to the subject of this volume. Also included is information on the samples collected, the location and method of collection, how the sample was analyzed, and other explanatory comments by the authors of this volume. See the Introduction Section for an explanation of the arrangement of data and for a general discussion of the data.

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Sp. 22, Sing

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UNCLASSIFIEDRADIONUCLIDE COMPOSITION
AIR OR BALLOON, RANGER

OPERATION RANGER, JANUARY 1951, NEVADA

Ref. 10, Spence
Ref. 18, Singlevich
Ref. 34, Spence
Ref. 44, Singlevich

Able	(1;A)	1.3 kt	1060 ft
Baker-1	(2;B ₁)	7.4 kt	1080 ft
Easy	(3;E)	1.0 kt	1080 ft
Baker-2	(4;B ₂)	7.7 kt	1100 ft
Fox	(5;Freddy;F)	22.0 kt	1435 ft

selected
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method of
tory comments
on for an
discussion of

Ref. 10 discusses various aspects of weapons development and includes radiochemical results of Operation Ranger. Samples of post-shot bomb debris were collected, beginning about one hour after each shot. Air-filtering units installed on manned B-29 bombers were used to collect the samples. Two types of filter paper were used on Shot Able; one (labeled 1) was developed by the Institute of Paper Chemistry (IPC), the other (labeled 2) was a Chemical Corps Type V filter. Only IPC filter papers were used for subsequent collections of the operation.

After the organic material of the papers was decomposed by digestion with acids, any silica remaining was removed by centrifugation and was dissolved. The silica solution and the supernatant from the centrifugation were combined and made up to 100-ml volume stock solution, from which aliquots were taken for radiochemical analyses. If necessary, dilutions were made for the analysis of high-yield fission products.

For the determination of the fraction of the bomb represented by a sample, it was necessary to know the fraction of the fissions occurring in the fissile material. This value was difficult to determine since fission yields are sensitive to the energy of the neutrons causing fission. Not much was known about the neutron spectrum existing when a bomb detonates except that the neutrons are fast. To provide calibration values of the fission yields, experiments were performed which simulated bomb neutrons in two different ways: The first was to use port 5W in the Fast Reactor at Los Alamos, which gave neutrons which were fast but somewhat degraded from original fission neutrons; the second approach was to irradiate samples in a capsule of ²³⁵U whose walls were thick enough to stop all thermal neutrons. The sample saw only fast neutrons from fissions in the capsule wall. It was observed that the values obtained in the case of the capsule irradiations depended on the position of the capsule in the thermal column. Evidently some nonthermal neutrons were leaking through the capsule walls. Accordingly, the capsule position was standardized as far out in the thermal column as was feasible. This position was 47 in. from the reactor face. The activity-ratio values due to bomb neutrons were expected to fall between the values determined at the 5W and the capsule locations.

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SECTION III LIST OF REFERENCES

The references listed in Section III are the sources of the data included in the Compendium of Data for Part 2 of Volume IV. As described in the Introduction, the bibliographical entries have been brought up to date since the first listing in Volume I of the Series. A few new ones have been added.

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13. ABSTRACT <p>Many technical reports have been published that contain data describing local fallout from nuclear weapon tests. Data on fallout from dissimilar types of weapons detonated under different conditions usually are not directly comparable, and thus far only limited attempts have been made to summarize these data so as to characterize local fallout. Correlating a given body of data would require that it be converted to a standard form, the significant parameters be computed, and these be compared according to some theory or empirical formula. A prerequisite would be the compilation of the data and authoritative commentary. For this purpose, a study was made that resulted in five volumes of such data, published as a series under the general title <u>Local Fallout from Nuclear Test Detonations</u>.</p> <p>The study was intended to critically select and compile, from published reports, data on all characteristics of local fallout from nuclear weapon tests held through 1958. These reports would be selected from the series of reports published prior to the first moratorium on above-ground test detonations. However, the Dominic Test Series of 1962 extended the time span to be covered; thus, the first four volumes contain the data for tests through 1958. The last volume of the Series includes material published on weapon tests conducted through 1962.</p> <p>The first volume in the series, <u>Indexed Bibliography of United States and British Documents on Characteristics of Local Fallout</u>, was published in 1961. It made available a ready reference to 206 reports from which relevant information was to be selected, while the selection and compilation of data progressed. The volume consists of an index to the references by fallout characteristics, an index to the references by authors' names, and a list of the references.</p>		

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13. ABSTRACT <p>The second volume, <u>Compilation of Fallout Patterns and Related Test Data (U)</u>, was published in several parts in 1963-66. It contains information on fallout patterns, on wind and cloud data, and on all that is pertinent to shot conditions, such as yield, burst height and placement.</p> <p>The third volume, <u>Annotated Compendium of Data on Physical and Chemical Properties of Fallout (U)</u>, was published in 1966. It contains the more significant of the data published that pertains to the physical and chemical nature of local fallout particles. Included are general statements made by the original investigators on the physical and chemical nature of the particles, as well as specific observations produced by detailed experimental analyses. Also included wherever possible are data descriptive of the samples analyzed, together with relevant information on collection equipment, location of collection, processing involved, and purpose of the analyses.</p> <p>This, the fourth, volume is a compendium of data on radiation and radiochemical characteristics of local fallout. It is published in two parts. Part 1 presents data on radioactivity investigated as a function of mass, particle size, and time. Accordingly, Part 1 is organized into three main data sections: Specific Activity, Activity-Size Distribution, and Decay. This part, Part 2, presents data relating activity measurements with radiochemical content (sections on Radionuclide Composition and Induced Activity) and energy (section on Gamma Spectra).</p> <p>Again, as in the third volume, included besides the primary data is background information on the location and types of collection equipment used, the instrumental or radiochemical processing involved, and the purposes of the analyses.</p>	

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13 ABSTRACT The fifth volume, Transport and Distribution of Local (Early) Fallout From Nuclear Weapon Tests (U), was published in 1965. It presents and evaluates information relating to nuclear detonations through 1962, that concerns the formation of the particle cloud, particle fall behavior, fallout dynamics, and fallout pattern features.		

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